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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF	:
RALF MAUS, ET AL.	: EXAMINER: NGUYEN, NGOC YEN M.
SERIAL NO: 10/067,841	:
FILED: FEBRUARY 8, 2002	: GROUP ART UNIT: 1754
FOR: PRECIPITATED SILICAS HAVING A NARROW PARTICLE SIZE DISTRIBUTION	:

DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

Now comes Dr. Ralf Maus, who deposes, and states that:

1. I am a graduate of Chemical Engineering and received my doctoral degree in the year 1996.

2. I have been employed by Degussa AG, Germany for 7 years as a Process Engineer in the field of Process Technology and Engineering.

3. The following experiments were carried out by me or under my direct supervision and control.

Figures 6 to 8, of the present application, Application No. 10/067,841, show precipitated silica dried using a pulse combustion dryer (PCD). To show the differences between the PCD-dried silica and silica dried according to conventional drying mechanisms, as used in U.S. Patent 6,383,280 and U.S. Patent 6,180,076, the following comparison experiments were preformed.

In one study, a comparison experiment was carried out using an analogue silica slurry as used for the PCD-dried silica depicted in Figures 6 to 8 of the invention. However, different drying mechanisms, a spray dryer and a "floor dryer with subsequently grinding," were used to dry the precipitated silica, instead of a pulse combustion dryer. The resulting silica particles were examined by scanning electron microscopy (Figures A-C and D-F).

In a second study, silica particles were prepared according to Example 4, as disclosed in U.S. 6,180,076. In accordance with Example 4, the silica was dried in a short time dryer, and the resulting silica particles were examined by scanning electron microscopy (Figures G-I).

In a third study, size distribution data for PCD-dried silica was compared to silica dried by a spray dryer and a "floor dryer with subsequently grinding," and with initial silica suspensions.

The scanning electron micrographs obtained from both studies are described as follows.

Figures A-C represent scanning electron micrographs of precipitated silica dried in a spray dryer.

Figures D-F represent scanning electron micrographs of precipitated silica dried in floor dryer and subsequently milled.

Figures G-I represent scanning electron micrographs of silica prepared according to Example 4 of U.S. 6,180,076, as powder, after short time drying.

Figures J-L represent scanning electron micrographs of silica prepared according to Example 4 of U.S. 6,180,076, as granule, granulated after a short time drying.

In the third study, a bar graph, Figure M, depicting particle size distributions, is provided, as shown below.

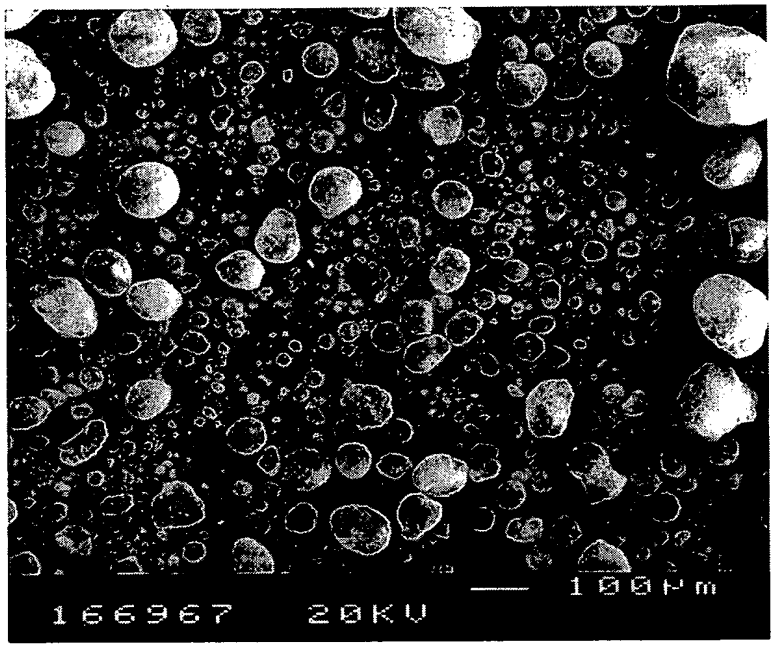

Figure A: Scanning electron micrograph of precipitated silica dried in a spray drier

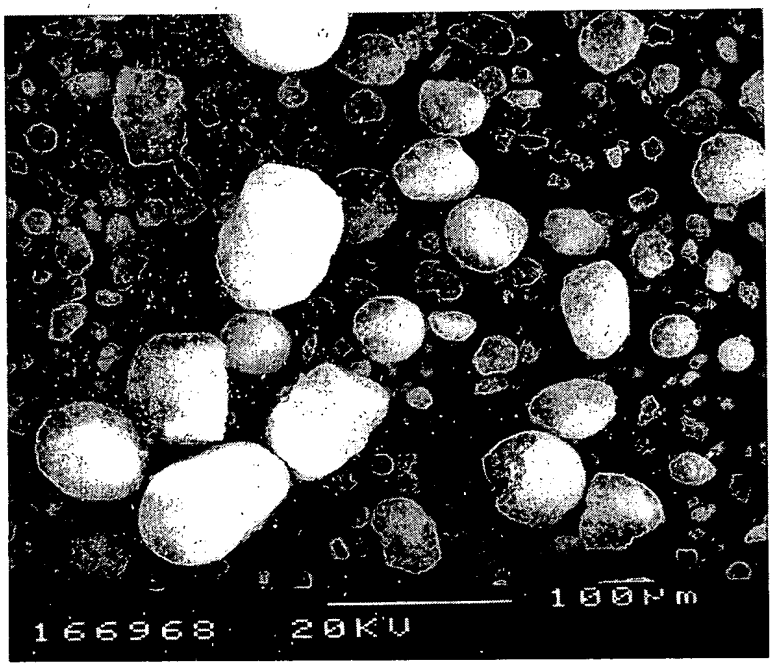

Figure B: Scanning electron micrograph of precipitated silica dried in a spray drier

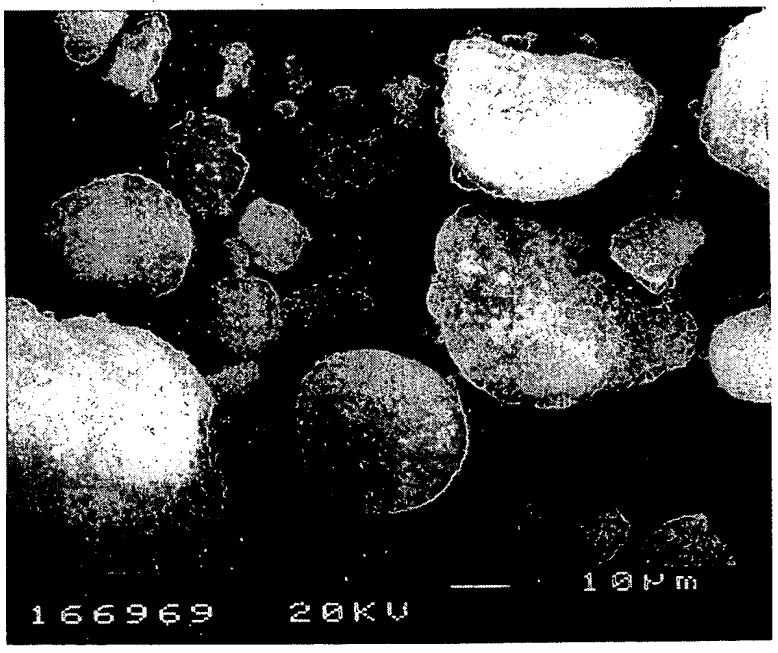

Figure C: Scanning electron micrograph of precipitated silica dried in a spray drier

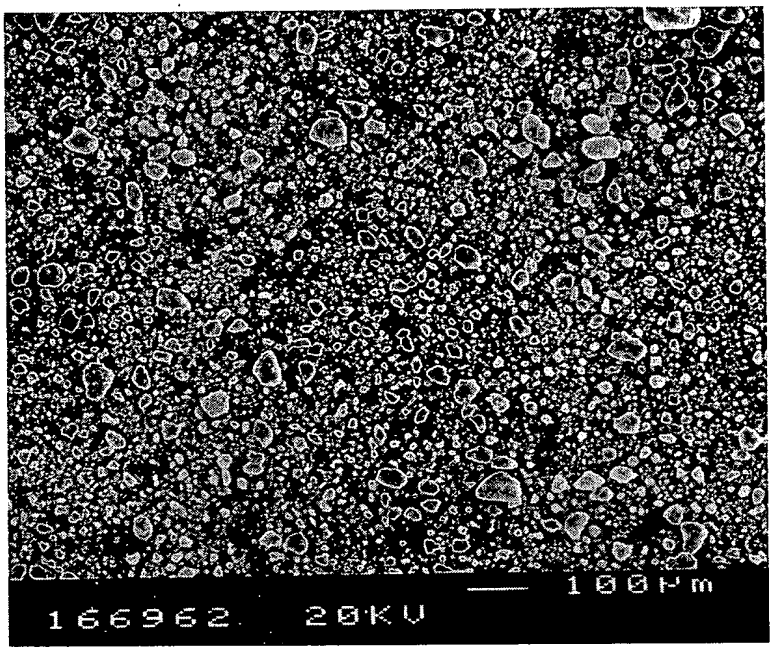

Figure D: Scanning electron micrograph of precipitated silica dried in a floor drier and subsequently milled

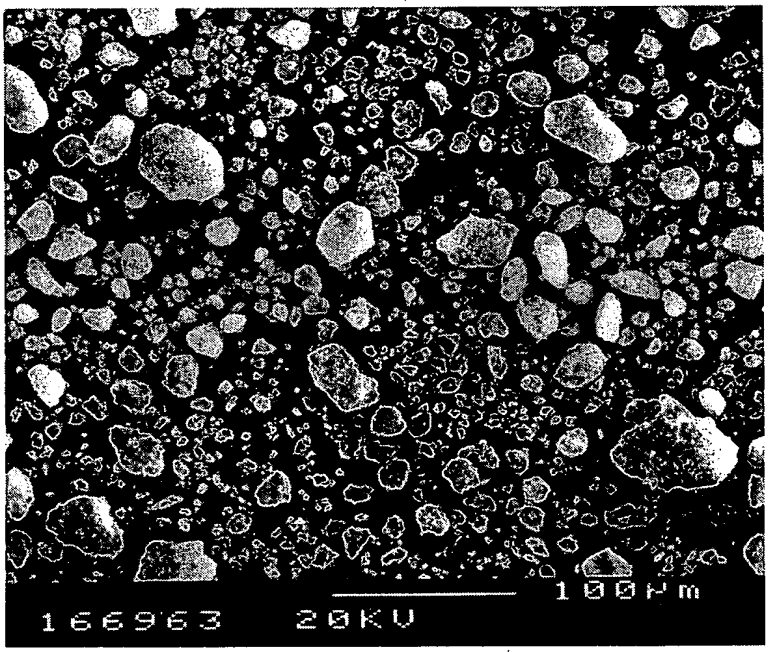

Figure E: Scanning electron micrograph of precipitated silica dried in a floor drier and subsequently milled

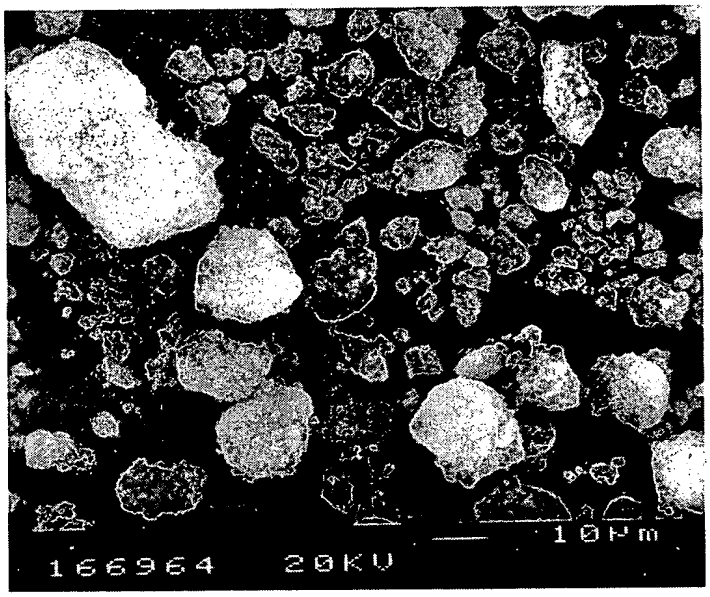

Figure F: Scanning electron micrograph of precipitated silica dried in a floor drier and subsequently milled

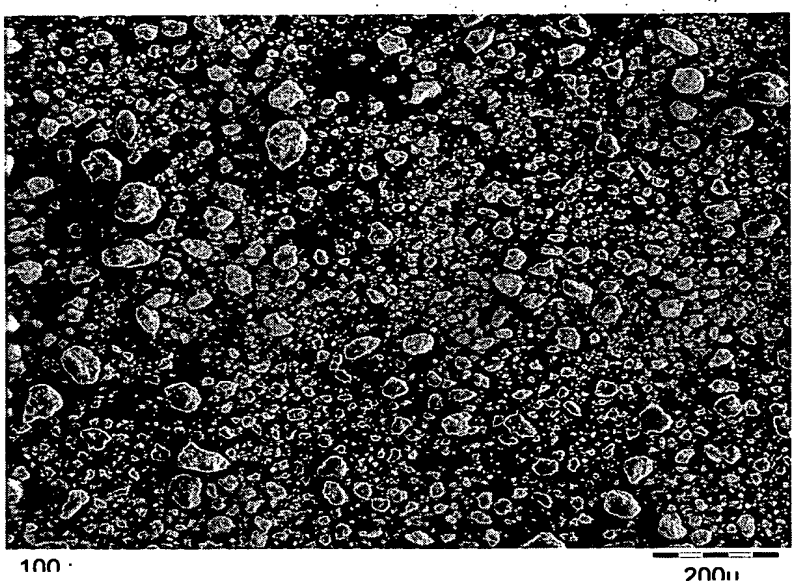

Figure G: Scanning electron micrograph of the silica according to Example 4 of U.S. 6,180,076 as powder after short time drying

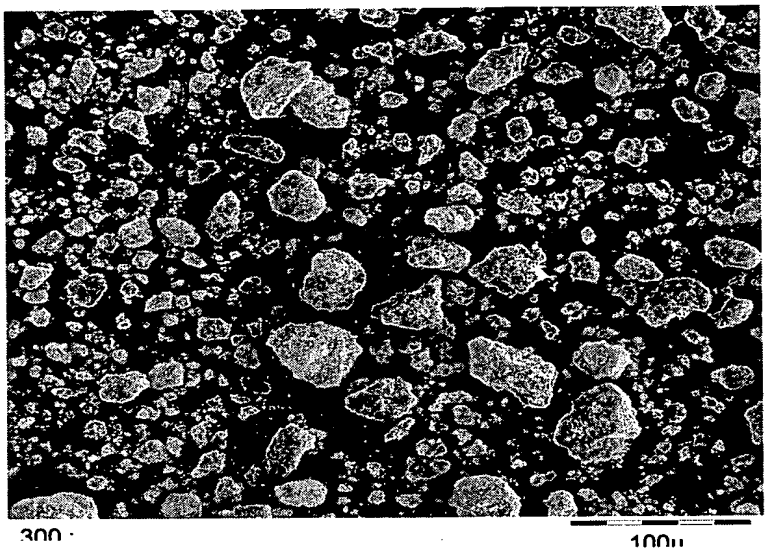

Figure H: Scanning electron micrograph of the silica according to Example 4 of U.S. 6,180,076 as powder after short time drying

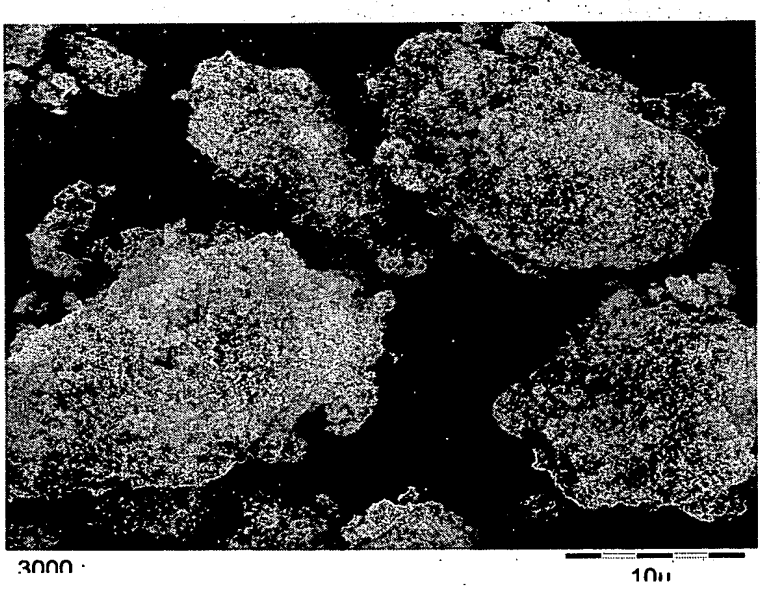

Figure I: Scanning electron micrograph of the silica according to Example 4 of U.S. 6,180,076 as powder after short time drying

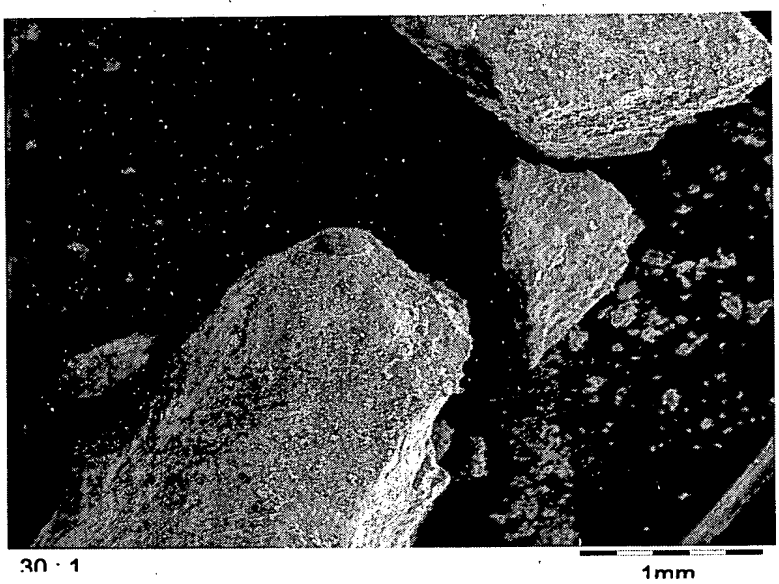

Figure J: Scanning electron micrograph of the silica according to Example 4 of U.S. 6,180,076 as granule, granulated after short time drying

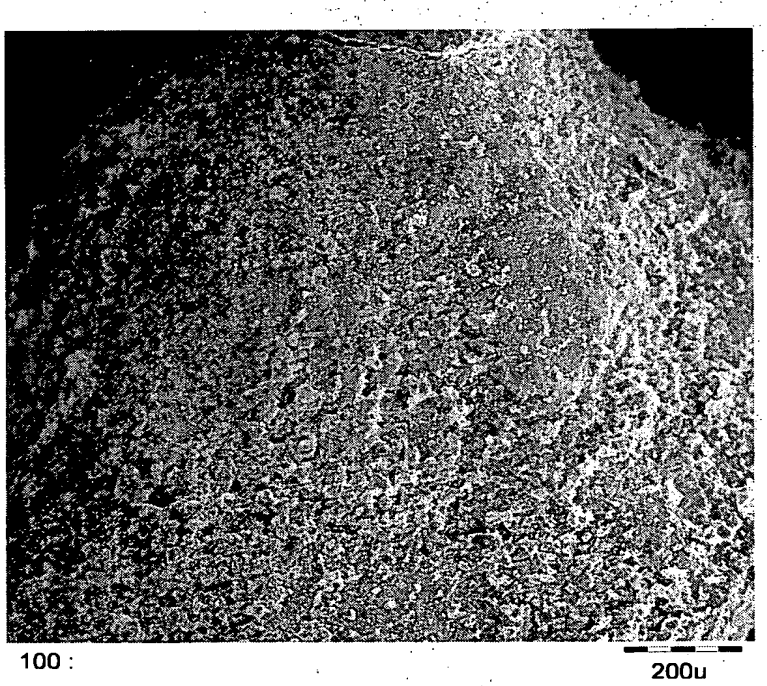

Figure K: Scanning electron micrograph of the silica according to Example 4 of U.S. 6,180,076 as granule, granulated after short time drying

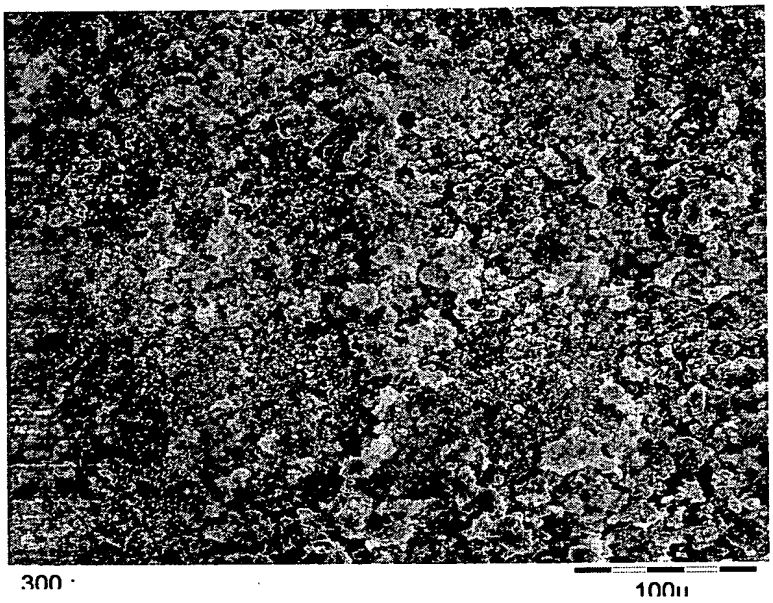

Figure L: Scanning electron micrograph of the silica according to Example 4 of U.S. 6,180,076 as granule, granulated after short time drying

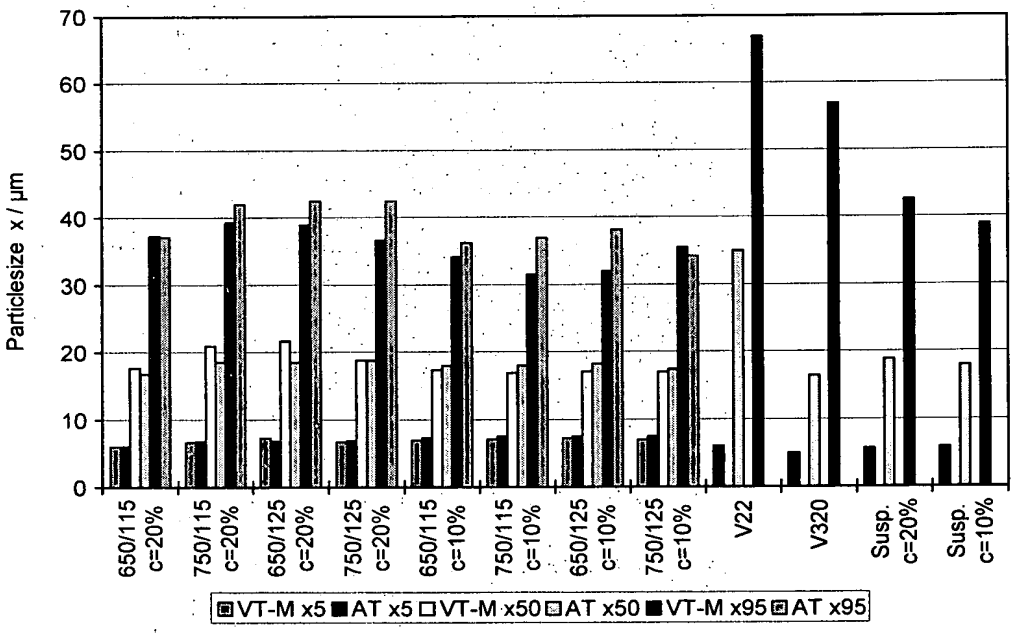

Figure M: Particle size distributions for PCD dried silica (cyclone), spray dried silica, floor dried and ground silica and silica suspensions

In the first study, precipitated silica dried in a spray drier is depicted in Figures A-C, and precipitated silica dried in a floor drier and subsequently milled is depicted in Figures D-F. A comparison of Figures 6 to 8 of the invention with Figures A to F, confirms that the silica of the invention, dried with PCD, exhibit a more uniform spherical shaped body and a significantly narrower particle size distribution. (see page 5, line 31 to page 6, line 9 of the present specification). The surface of the spray dried and floor dried silica (Figures C and F) are grossly irregular in form, indicating the possible formation of hollow spheres. However, no indication of formation of hollow spheres is observed in the PCD-dried silica (see also page 6, lines 6-7 of the present specification). In addition, the silica of the invention exhibits a smoother particle surface, and has virtually nearly no irregularities, as demonstrated by Figure 8 in comparison with Figures C and F. This comparison study confirms that silica particles obtained by PCD differ from silica particles obtainable by classical drying technologies, such as spray drying and floor drying.

In the second study, silica prepared according to Example 4 of U.S. 6,180,076, as powder after short time drying is depicted in Figures G-I. This patent also discloses granulated products, thus the powder depicted in Figures G to I, was granulated. Pictures of the resulting granules are depicted in Figures J to L. This patent also discloses ground (milled) silica, which is comparable in form to the silica particles depicted in Figures D to F, above.

In the second study, a comparison of Figures 6 to 8 of the invention with Figures G to L, confirms that the silica of the invention, dried with PCD, exhibit a more uniform spherical shaped body and a significantly narrower particle size distribution than these corresponding features of silica powder prepared by Example 4 of U.S. 6,180,076, and such particles prepared by subsequent granulation or milling (ground silica). In addition, the silica of the invention

exhibits a smoother particle surface, and has virtually nearly no irregularities, as demonstrated by Figure 8 in comparison with Figures I and L. This comparison study confirms that silica particles obtained by PCD differ from silica particles obtainable by short time drying of U.S. 6,180,076, and such particles prepared by subsequent granulation or milling.

In addition to the studies presented above, a third study, provided below, compares the results of Figure 12 (PCD silica) of the invention with new distribution data, shown in Figure M.

Figure 12, of the invention, provides the d_5, d_{50} and d_{95} values of the initial silica suspensions (4% and 8%, see bar graph groupings at positions 1-2), as well as for eight silica preparations dried with PCD under different apparatus set-ups (split into cyclone and filter product each; see bar graph groupings at positions 3 -18). The following tables are provided from an analysis of Figure 12.

A) Particle size distribution for 4% slurry

	Initial Slurry	Cyclone (main product)	Filter (side product)
d_5 [µm]	3	4-4.5	2-3
d_{50} [µm]	9	7-8	3-5
d_{95} [µm]	21	18	8-33

B) Particle size distribution for 8% slurry

	Initial Slurry	Cyclone (main product)	Filter (side product)
d_5 [µm]	3	3.5-4	2
d_{50} [µm]	9	7-8	3-5
d_{95} [µm]	20	17-18	5-33

The particle size distribution of the initial slurry ranges from 3 to 21µm, while the particle size distribution of the main product (cyclone) ranges from 3.5 to 18µm.

Consequently, the particle size distribution of the PCD-dried silica (cyclone) is narrower than that of the initial slurry.

Figure M compares the particle size distributions of particles prepared from a silica slurry preparation, as used with the PCD-dried silica, and dried by spray drying or "floor drying and grinding," with PCD-dried particles of the invention, and with silica suspensions.

In Figure M, bar graph groupings at positions 1 to 8 correspond to PCD-dried silica (cyclone), bar graph groupings at position 9 correspond to spray dried silica, bar graph groupings at position 10 correspond to floor dried and ground silica, and bar graph groupings at positions 11 and 12 correspond to the silica suspensions. Each column includes d_5, d_{50} and d_{95} values, and bar graph groupings at positions 1 to 8 comprise double determinations of all three values. The PCD experiments displayed in bar graph groupings at positions 1 to 8 were carried out employing different apparatus set-ups. The following table results from an analysis of Figure M.

C) Particle size distribution – Figure M

	Initial Slurry	PCD-dried	Spray Dried	Floor Dried + Ground
d_5 [μm]	5	6-7	6	5
d_{50} [μm]	18-19	17-22	35	16
d_{95} [μm]	38-42	31-42	68	57

It is apparent that the particle size distribution of the silica product dried with PCD is similar, to or narrower than, that of the initial suspension, and significantly narrower than the particle size distribution for each of the particle size distributions of the silica particles obtained by spray drying and "floor drying and grinding." In contrast to the PCD-dried silica, the particle size distributions of the silica particles obtained by spray drying and "floor drying and grinding" are each much broader than the distribution of the initial suspension. These results confirmed, that a pulse combustion drying mechanism leads to silica particles

exhibiting improved particle size distributions, as compared to the spray drying and "floor drying and grinding" mechanisms, and better or comparable distributions compared to initial silica suspensions.

4. I declare further that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true, and further that these statements were made with knowledge that willful false statements, and the like, so made, are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

5. Further Declarant saith not.

Respectfully submitted,

10/12/04
Date